

REPORT DOCUMENTATION PAGE				Form Approved OMB No. 0704-0188	
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1. REPORT DATE (DD-MM-YYYY) 12-14-2012		2. REPORT TYPE FINAL		3. DATES COVERED (From - To) 09/05/2009 - 09/14/2012	
4. TITLE AND SUBTITLE CARBON NANOTUBE QUANTUM DOTS AS THZ DETECTORS				5a. CONTRACT NUMBER	
				5b. GRANT NUMBER FA9550-09-1-0697	
				5c. PROGRAM ELEMENT NUMBER	
6. AUTHOR(S) Paola Barbara				5d. PROJECT NUMBER	
				5e. TASK NUMBER	
				5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Georgetown Nanoscience and Microtechnology Laboratory (GNuLab) Department of Physics Georgetown University 37th St. and O St. NW Washington, DC 20057-1228				8. PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) Air Force Office of Scientific Research/RSE Suite 325, Room 3112 875 Randolph Street Arlington, VA 22203-1768				10. SPONSOR/MONITOR'S ACRONYM(S) AFOSR	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S) AFRL-OSR-VA-TR-2012-1239	
12. DISTRIBUTION / AVAILABILITY STATEMENT Distribution A - Unlimited					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT In this project we developed very sensitive broadband THz detectors using carbon nanotube quantum dots coupled to antenna-shaped source and drain electrodes. The sensing mechanism is photon-assisted tunneling and leads to a counterintuitive effect: the THz irradiation reduces the effective temperature of the electrons in the dot and drastically increases the height of the Coulomb blockade peaks, greatly enhancing the device performance. This type of detector is very attractive because 1) it is highly sensitive (THz power in the femtowatt range can be easily detected), 2) unlike other THz detectors such as bolometers, it has a frequency dependent response, with spectral resolution within about a tenth of THz via photon assisted tunneling, that is inelastic tunneling of single electrons through the dot via absorption or emission of photons. We also find a strongly enhanced response that orthodox theories of photon assisted tunneling fail to quantitatively explain. We suggest that this effect is due to tunneling through excited states and non-equilibrium cooling in the presence of THz radiation.					
15. SUBJECT TERMS					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT UU	18. NUMBER OF PAGES 8	19a. NAME OF RESPONSIBLE PERSON Dr. Harold Weinstock
a. REPORT U	b. ABSTRACT U	c. THIS PAGE U			19b. TELEPHONE NUMBER (include area code) 703-696-8572

Grant/Contract Title: CARBON NANOTUBE QUANTUM DOTS AS THZ DETECTORS

In this project we developed very sensitive broadband THz detectors using carbon nanotube quantum dots coupled to antenna-shaped source and drain electrodes. The sensing mechanism is photon-assisted tunneling and leads to a counterintuitive effect: the THz irradiation *reduces* the effective temperature of the electrons in the dot and drastically increases the height of the Coulomb blockade peaks, greatly enhancing the device performance. This type of detector is very attractive because 1) it is highly sensitive (THz power in the femtowatt range can be easily detected), 2) unlike other THz detectors such as bolometers, it has a frequency dependent response, with spectral resolution within about a tenth of THz via photon assisted tunneling, that is inelastic tunneling of single electrons through the dot via absorption or emission of photons. We also find a strongly enhanced response that orthodox theories of photon assisted tunneling fail to quantitatively explain. We suggest that this effect is due to tunneling through excited states and non-equilibrium cooling in the presence of THz radiation.

In the future we plan to extend this work to graphene to probe new physics in this material and open the way to carbon-based THz devices that are viable for wafer-scale production. We recently started testing fabrication processes using epitaxial graphene on SiC in collaboration with Dr. Gaskill at the Naval Research Laboratory.

Publications acknowledging support from this project are in refs. [1-3].

Below is a summary of the work carried out under this grant.

Background on quantum dots and photon assisted tunneling.

A short section of carbon nanotube or graphene nanoribbon that is weakly coupled to source and drain electrodes via high resistance contacts behaves as a quantum dot. Electrons tunnel in the dot one at a time and a substantial charging energy U is needed to add a single electron to the dot ($U = e^2/C$, where C is the total capacitance of the dot) [4]. Therefore, the energy spectrum of

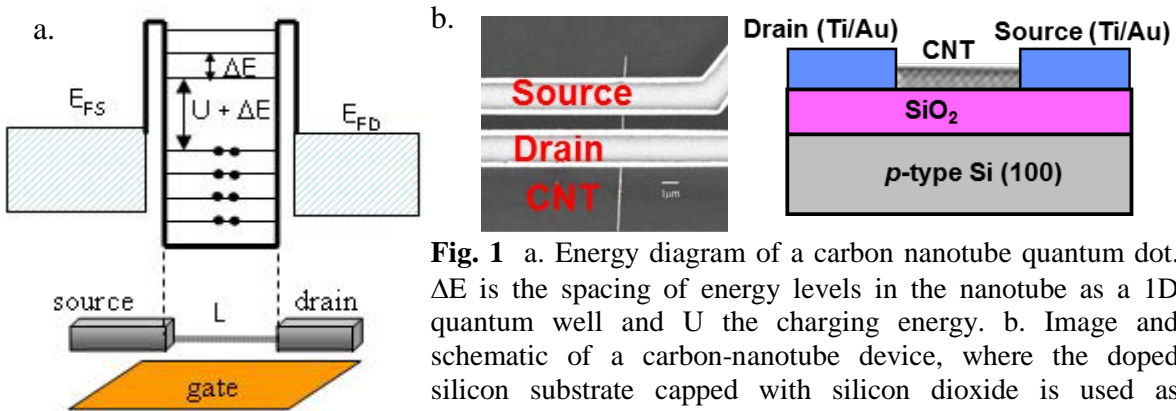


Fig. 1 a. Energy diagram of a carbon nanotube quantum dot. ΔE is the spacing of energy levels in the nanotube as a 1D quantum well and U the charging energy. b. Image and schematic of a carbon-nanotube device, where the doped silicon substrate capped with silicon dioxide is used as backgate.

the electrons in the dot is characterized by the charging energy U and the energy level spacing ΔE , which is inversely proportional to length L of the carbon nanotube or nanoribbon section, according to $\Delta E = \hbar v_F \pi / L$ ($\Delta E = 1.67$ meV for $L = 1$ μm). The charging energy opens a gap at the Fermi energy (see Fig. 1a). A voltage applied to a nearby gate electrode shifts the Fermi energy in the dot, therefore the source-drain current as a function of gate voltage shows sharp peaks, corresponding to resonant elastic tunneling of electrons, one at a time, occurring when the Fermi

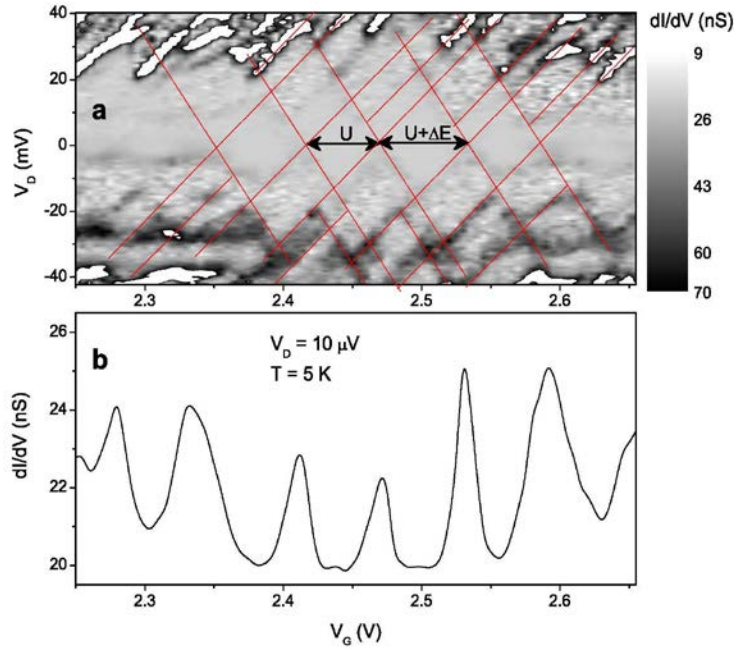


Fig. 2: a. Differential conductance (grayscale) as a function of source-drain and gate voltage for a carbon nanotube quantum dot with on-chip antennas and top gate. The gate dielectric is PMMA. The diamonds of different sizes are due to spin degeneracy. b. Coulomb peaks of conductance vs. gate voltage at fixed source-drain voltage. [3]

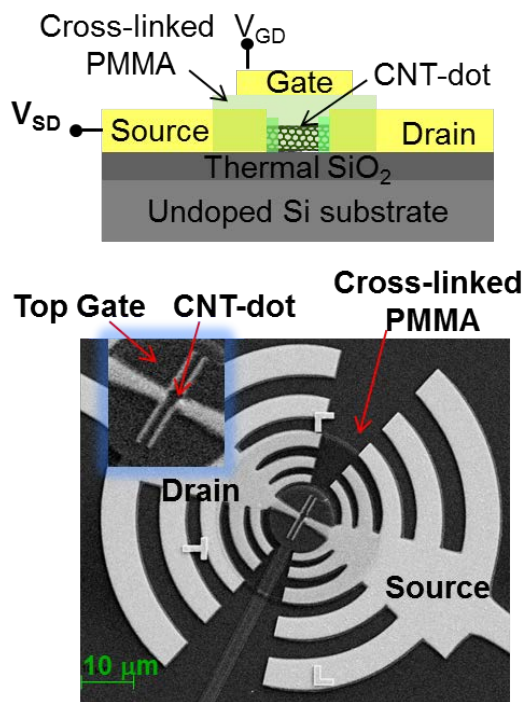


Fig. 3: Schematic (top) and SEM image (bottom) of a carbon nanotube quantum dot with on-chip antenna and top gate. [3]

energies of the leads are aligned with an energy level in the dot (see Fig. 2b). Coulomb blockade occurs between the peaks and the voltage interval between adjacent peaks is characterized by the energy level spacing and the charging energy of the dot [4]. When an electromagnetic field is present, transport can also occur via photon assisted tunneling, that is inelastic tunneling with absorption or emission of photons [5]. Photon assisted tunneling generates side peaks in the Coulomb blockade regions: the voltage interval between the side peak and the main peak is determined by the photon energy ($hf \cong 4$ meV at 1 THz) and the height of the side peaks is related to the intensity of the electromagnetic field [5-7]. Carbon nanotubes and graphene nanoribbons are ideal quantum dots for photon assisted tunneling in the THz range, because their typical charging energy ($E_C \sim 10$ meV or larger for lengths smaller than a micron) is at least one order of magnitude larger than typical charging energies obtained for other types of quantum dots (e.g. 2DEG of GaAs/Al_xGa_{1-x}As

heterostructures) allowing detection of side peaks up to a few terahertz, at power levels on the order of femtowatts. The lower frequency that can be detected is comparable to the width of the peaks, which depends on the temperature and on characteristic quantum dot properties, such as the coupling between the nanotube and the leads. All the characteristic energies of the device can be extracted from the diamond shapes in the plots of conductance as a function of gate voltage and bias, as shown in Fig. 2a for one of our devices.

Reports of photon-assisted tunneling in quantum dots are scarce and the effect is largely unexplored at frequencies higher than tens of GHz. This is mainly due to the difficulty to efficiently couple THz radiation to the devices and the limited availability of powerful THz sources. In our AFOSR sponsored project, we have introduced **on-chip antennas** to efficiently couple THz radiation to carbon nanotube devices and effectively study the interaction between the quantum dots and the THz radiation [3]. Details of our design will be discussed in the section titled “Summary of results”. Understanding general properties of carbon nanotube devices, specifically hysteresis and how to reduce its effects on our detectors was an important preliminary part of our project [1, 2].

Quantum dot THz detectors vs. bolometers.

Graphene THz detectors based on a different detection method, bolometric response, have also been successfully developed [8]. A significant drawback however, is that the bolometric response does not provide spectral resolution, whereas, due to its quantum nature, photon assisted tunneling in quantum dots yields a frequency dependent response. Another drawback is that, because the bolometer detector makes use of the temperature dependence of the resistivity of gapped bilayer graphene, it is a low temperature device. The quantum dot detectors proposed here also work at low temperature, but we can push their operation to higher temperatures by fabricating devices with higher charging energies. Carbon nanotubes single-electron transistors operating at room temperature have in fact been demonstrated, with a combination of charging energy and electron level spacing as high as 120 meV [9].

Carbon nanotube quantum dots THz detectors.

I. Kawano *et al.* [10] recently demonstrated detection via photon assisted tunneling for a carbon nanotube quantum dot structure similar to the one showed in Fig. 1b (silicon substrate as back-gate, no antennas). The device was irradiated with four gas laser sources at four different

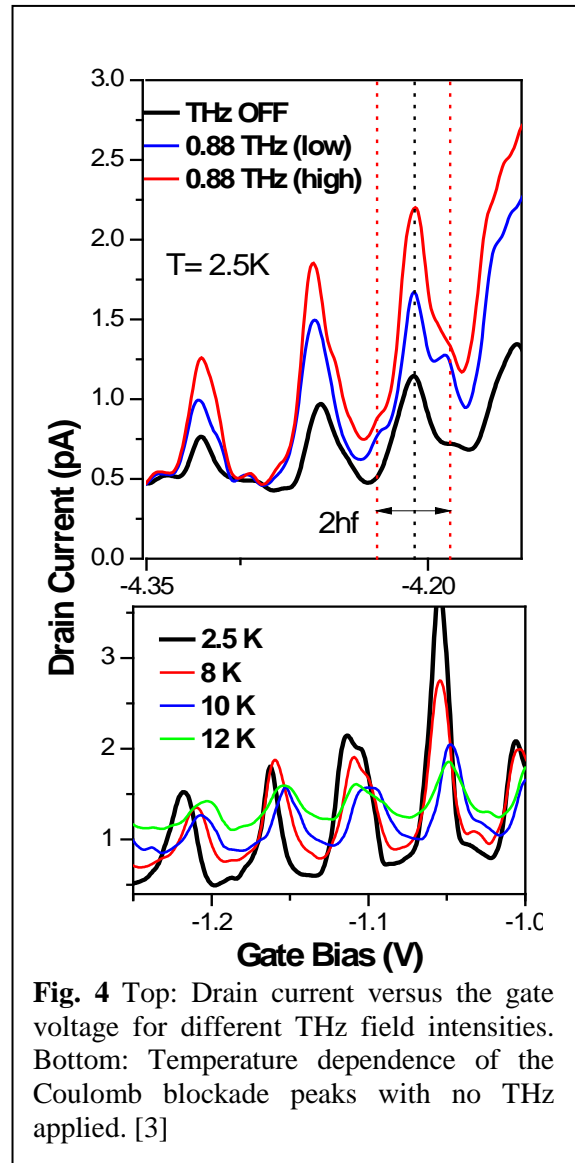


Fig. 4 Top: Drain current versus the gate voltage for different THz field intensities. Bottom: Temperature dependence of the Coulomb blockade peaks with no THz applied. [3]

frequencies [10] and satellite peaks were measured with spacing from the main peak proportional to the frequency. Notwithstanding this report of successful detection, two main challenges hampered further research. The first was the small coupling between the quantum dot and the powerful (10 mW) laser source. The second was the difficulty to reproduce the results on multiple samples. Among many devices tested, only very few responded to external radiation [11].

Summary of results.

Efficient coupling of THz radiation to quantum dots requires a careful design of the device architecture. In addition to high reflection losses due to the mismatch between the detector impedance and the free space impedance, the dimension of the effective detection region (which is comparable to the size of the quantum dots, about ~ 300 nm) is much smaller than the radiation wavelength (300 microns at 1 THz in free space), as well as the region in which the THz power can be focused (~ 1 mm²). On-chip antennas are therefore necessary to couple THz radiation to carbon nanotube devices. Antennas significantly increase the effective detection area and reduce impedance mismatch. Since we test the device response as a function of frequency, we select antenna designs that work in a wide frequency range.

Planar self-complementary antennas [12] are good candidates because their impedance is frequency independent and close to the impedance of free space (within about a factor of two). One example is shown in Fig. 3, where the source and drain electrode are patterned as log-periodic antennas [13], designed to operate in the frequency range 700 GHz to 2.5 THz. For these samples, the substrate is intrinsic silicon and the gate electrode is patterned on the top, after growing an insulating layer on the device (the carbon nanotube connecting the source and drain electrode is underneath the small circular top gate electrode). In our samples we used cross-linked PMMA as a dielectric layer. Although these quantum dots do not show features that are as sharp as they are for quantum dots with doped Si backgate and SiO₂ as dielectric, we can still distinguish the diamond pattern and extract the quantum dot parameters ($U = 13.5$ meV, $\Delta E = 3.5$ meV for the quantum dot in Fig. 2).

We successfully detected THz radiation from quantum dot devices with on-chip antennas by using two types of THz sources [3]. The first is a backward wave oscillator BWO source, with frequency doubler and tripler. The BWO tunable

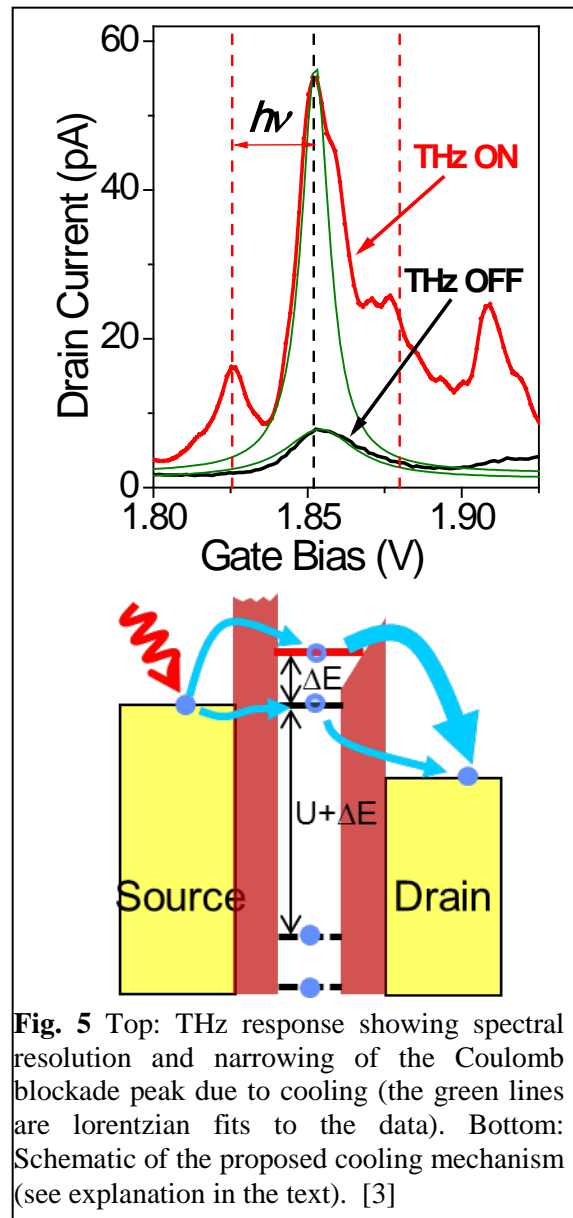


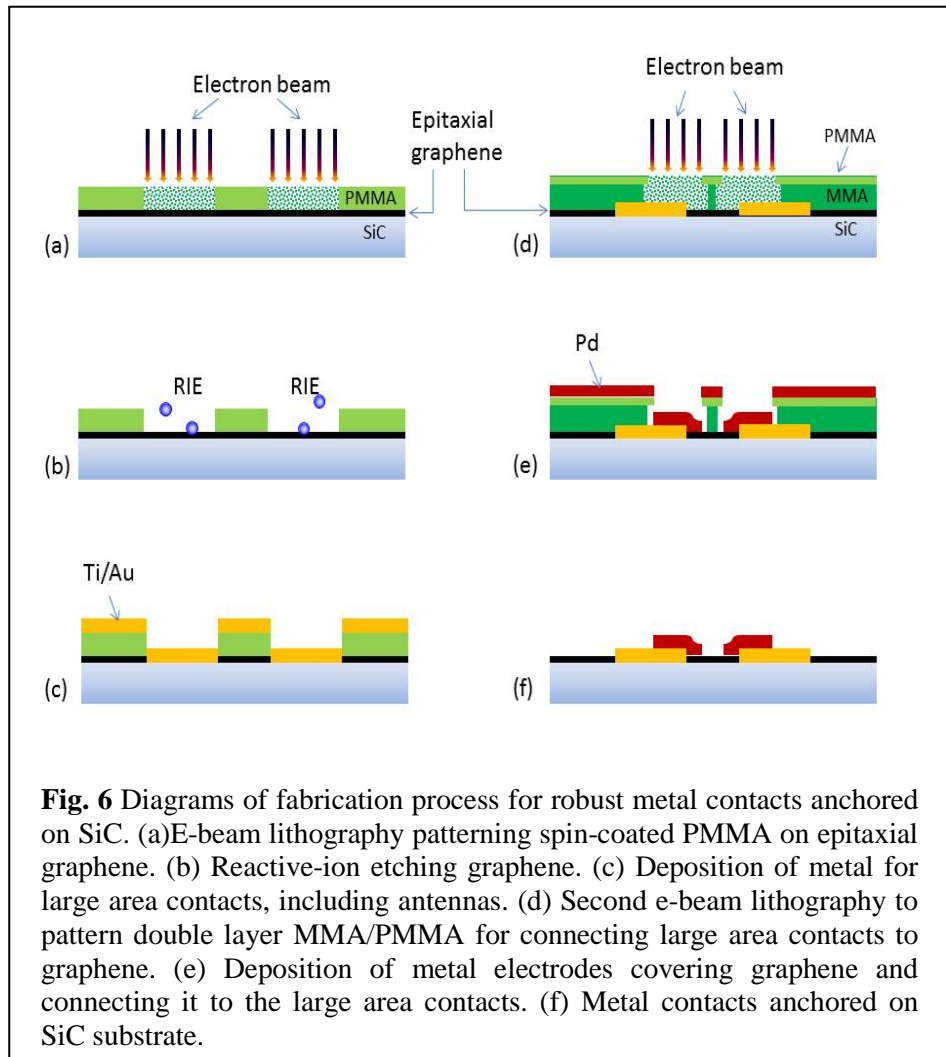
Fig. 5 Top: THz response showing spectral resolution and narrowing of the Coulomb blockade peak due to cooling (the green lines are lorentzian fits to the data). Bottom: Schematic of the proposed cooling mechanism (see explanation in the text). [3]

output is from 600GHz to 1080 GHz and power ranges from 0.1 mW at low frequency to 0.02 mW at high frequency. The second is a laser source at the University of Maryland in Prof. Drew's laboratory. The frequencies of the available CO₂ pumped laser lines range from 0.7 THz to 7 THz.

The on-chip antennas provided a strong coupling between the carbon nanotube quantum dot and the external THz radiation. An example of the strong change in the current vs. gate voltage curves is shown in Fig. 4. Notably, the response cannot be quantitatively explained by using orthodox theories of photon assisted tunneling and Coulomb blockade. These theories predict that upon irradiation the main Coulomb blockade peak should decrease and the side peaks should increase, following a Bessel function dependence on the THz power [5-7]. However, the measurements show that the main peak increases and becomes narrower when THz radiation is applied. Such increase of the main peak is not caused by heating, since the Coulomb blockade peaks become much broader when the THz radiation is off and the sample temperature is raised, as shown in Fig. 4 (bottom graph). This anomalous response can be explained by non-equilibrium cooling of the dot. For sufficiently high frequencies, $hf > \Delta E$, photon assisted tunneling occurs through excited levels in the dot. If the tunneling rate from the dot to the drain electrode is higher for the excited level, the most energetic electrons will leave the dot at a faster rate and the electrons in the dot will cool down. The theoretical analysis from S. Shafraniuk shows that this cooling causes the sharp increase of the main peak [3]. Although for most experiments our operating temperature ranged from 2.5 K to 5 K, such strong response can still be detected when operating the device at higher temperature.

Graphene Quantum Dots THz detectors

We started testing how to extend the design successfully used for carbon



nanotubes to epitaxial graphene grown on SiC. In the case of carbon nanotubes, the fabrication of THz detectors described above is time consuming and it is not suitable for large-scale production. This is because carbon nanotubes are grown by chemical vapor deposition from catalyst island. This process requires first the patterning of catalyst islands, then the growth of

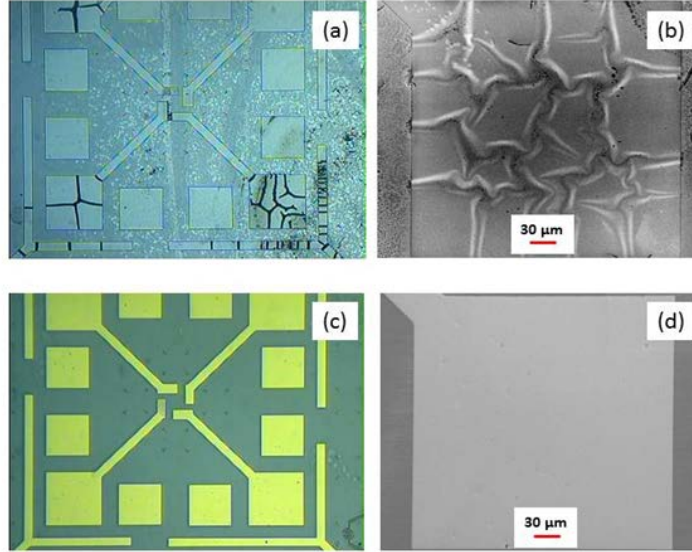


Fig. 7 (a) Optical image of Al pads and leads patterned by e-beam lithography where metal is deposited directly on epitaxial graphene (EG). (b) FESEM image of an Al pad on EG prepared the same way as (a). (c) Optical image of Ti/Au pads and leads patterned by e-beam lithography where metal is deposited after removing underneath graphene by RIE. (d) FESEM image of one of the Ti/Au pads in (c).

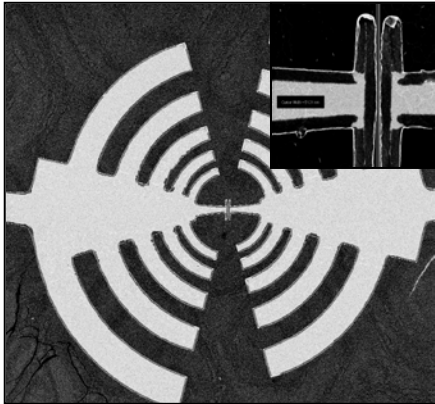


Fig. 8 Antenna patterned on a SiC substrate with epitaxial graphene. The gap between the electrodes is 400 nm. Crosslinked PMMA will be used to patter the graphene quantum dot between the electrodes.

carbon nanotubes and then the imaging of carbon nanotubes to find those suitable for the quantum dot fabrication. The next step requires alignment of electrodes, antennas and top gate to the specific carbon nanotube orientation. With epitaxial graphene grown on SiC all these steps will be avoided, because quantum dots can be created by etching graphene in any part of the substrate. This is a tremendous advance towards large scale device fabrication. However, there are other problems connected to device fabrication with epitaxial graphene on SiC. The first problem is that the graphene layer is easily contaminated by photoresist residues. The second is that the adhesion of metallic layers deposited on graphene is poor and leads to peeling problems for electrodes of large area such as our THz antennas.

Our proposed fabrication process to overcome these problems is outlined below and shown in Fig. 6. The

deposition of large area contacts is done directly on SiC, after etching the graphene in the areas

where the contact will be deposited. After depositing the contacts, we plan to pattern the graphene connecting the contacts in the shape of a quantum dot by using crosslinked PMMA as a mask. The crosslinked PMMA can be patterned to create dots as small as 10 nm. The graphene around the dot will be etched by reactive ion etching (RIE). The crosslinked PMMA will form part of the dielectric layer for the top gate. An additional layer of dielectric will be deposited before depositing the top gate, to finally obtain a graphene-based structure similar to the one we successfully developed for carbon nanotubes (see Fig.3).

Fig.7 and Fig. 8 show that the fabrication process described in Fig. 6 is successful in addressing the adhesion problem with the large metal contacts.

Once the large area pads and antenna are anchored to the substrate with good adhesion, the steps for deposition and patterning of the subsequent layers will be very similar to the process used for carbon nanotube quantum dots.

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